

# Suppressed compressibility at large scale in jammed packings of size disperse spheres

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We analyze the large scale structure and fluctuations of jammed packings of size disperse spheres, produced in a granular experiment as well as numerically. While the structure factor of the packings reveals no unusual behavior for small wavevectors, the compressibility displays an anomalous linear dependence at low wavevectors and vanishes when  $q \rightarrow 0$ . We show that such behavior occurs because jammed packings of size disperse spheres have no bulk fluctuations of the volume fraction and are thus hyperuniform, a property not observed experimentally before. Our results apply to arbitrary particle size distributions. For continuous distributions, we derive a perturbative expression for the compressibility that is accurate for polydispersity up to about 30%.

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When an assembly of hard particles is compressed, there comes a point where further compression is difficult because the required pressure is too large. A similar “jammed” state can be obtained with soft repulsive particles (as in emulsions or foams), at a particular volume fraction  $\phi_c$  above which the stability of the packing is controlled by the elasticity of the particles. A large body of recent work in both theory and experiment has characterized the properties of jammed packings [1, 2].

While much attention has focussed on contacts at the interparticle scale, and force networks and connectivity at larger scales [1–3], there has been comparatively little research into fluctuations and response at very large scales. In Ref. [4] the low-wavevector behaviour of the structure factor,  $S(q)$ , in a monodisperse system of hard frictionless spheres was studied numerically, revealing “unexpectedly” weak density fluctuations at low  $q$ ,

$$S(q) \approx aq, \quad (1)$$

for some constant  $a$ , as was also found later for soft particles [5]. This should be compared to the behaviour in liquids (including hard sphere fluids at  $\phi < \phi_c$ ), where  $S(q) \approx S(q \rightarrow 0) + a'q^2$  [6]. These results imply that bulk fluctuations in the number density are suppressed at  $\phi_c$ : in  $d$  dimensions, fluctuations of particle number in sub-systems of linear size  $L$  scale as  $\langle \Delta N^2 \rangle_L \sim L^{d-1}$ . Such suppressed density fluctuations are the defining feature of “hyperuniform” materials [8].

Two recent papers reported surprising results, failing to detect the behaviour in Eq. (1). A numerical simulation of binary mixtures [9] found that (1) only holds for the particular case of a monodisperse system. Similarly, a confocal microscopy study [10] of a jammed system of moderately polydisperse colloidal hard spheres also failed to observe Eq. (1). Both studies report that  $S(q)$  in size disperse systems is different from Eq. (1), and suggest that size disperse packings might not be hyperuniform.

That this is a highly topical question is demonstrated by the recent work of Ref. [11], where independently of our approach hyperuniformity was detected, using two-point probability functions.

Here, we report the first experimental observation of vanishing fluctuations of the volume fraction and hyperuniformity in a granular experiment. The same observation is made for numerically produced polydisperse packings with arbitrary size distributions. In contrast with [9, 10] we consider not only the structure factor  $S(q)$ , but also the isothermal compressibility,  $\chi_T(q)$ , the latter being central to our analysis of size disperse packings. We propose a novel perturbative approach for extracting  $\chi_T(q)$  for continuous size distributions, which in general is a non-trivial task, and explain the connection between vanishing compressibility and hyperuniformity.

First, we briefly describe our systems. Experimentally we produce dense random granular packings by slowly compressing horizontally vibrated bidisperse brass disks. Typically 4500 large disks (diameter  $5 \pm 0.025$  mm) and 3500 small disks (diameter  $4 \pm 0.025$  mm), surrounded by rigid walls, are placed on an oscillating glass plate (amplitude 5 mm, frequency 10 Hz). The packing fraction is increased logarithmically slowly ( $d\phi/d\log(t) \simeq 10^{-2}$ ) until the force  $F$  measured on the compressing wall increases sharply from  $F = 0.05 Mg$  to  $F > Mg$ , where  $M = 2$  kg is the typical total mass of the “grains”. At that point the packing jams, grains stop moving and the force measured at the wall remains finite when the vibration is switched off (see Ref. [13] for more details). We take a high resolution picture of the entire packing ( $2048 \times 2048$  pixels) at the largest packing fraction, correcting for optical distortion. We detect the positions of the grains with a resolution of  $20 \mu\text{m}$  and retain  $\approx 4000$  large grains and  $\approx 3000$  small grains at least 1 cm away from the walls. We analyze two independent packings, produced starting from uncorrelated initial conditions.

Rattler particles are always included in the analysis. Numerically we generate 3d polydisperse sphere packings at  $\phi_c$  using soft repulsive particles, as first proposed in [1], using conjugate gradient methods and small decompression steps to prepare packings exactly at  $\phi_c$  [1, 12]. For each set of parameters, we prepare a single, very large configuration composed of  $N = 64,000$  particles. We study both a 50:50 binary mixture of spheres with diameter ratio  $R \in [1, 2]$ , or systems with a continuous size distribution, which we take as a flat distribution centered around the average value  $\bar{\sigma}$ . We studied polydispersities up to  $p = 0.4$ , where  $p$  is the standard deviation of the size distribution divided by  $\bar{\sigma}$ .

We now provide some definitions. Consider a size disperse system composed of  $n$  species, containing  $N_i$  particles of species  $i$ , with diameter  $\sigma_i$ . We define  $N = \sum_{i=1}^n N_i$ , the density  $\rho = N/V$ , the concentrations  $x_i = N_i/N$ , and the partial density fields  $\rho_i(\mathbf{q}) = \sum_{j=1}^{N_i} e^{i\mathbf{q}\cdot\mathbf{r}_j}$ , where  $\mathbf{r}_j$  is the position of particle  $j$  belonging to species  $i$ . The partial structure factors read  $S_{ij}(q) = \frac{1}{N} \langle \rho_i(\mathbf{q}) \rho_j(-\mathbf{q}) \rangle$ , and we collect them in a matrix,  $\mathbf{S}(q)$ . The total structure factor is defined as usual:

$$S(q) = \sum_{i=1}^n \sum_{j=1}^n S_{ij}(q). \quad (2)$$

When we analyze  $S(q)$  in our experimental granular packings, see Fig. 1a, we find that the low- $q$  behaviour is not compatible with Eq. (1). The same observation holds, see Fig. 2a, for the evolution of  $S(q)$  for numerical packings with continuous size distribution of increasing polydispersity. The inset shows that  $S(q \rightarrow 0)$  increases continuously with the polydispersity  $p$ . This suggests that size dispersity is the main factor responsible for the numerical results in Ref. [9] and the experimental ones in Ref. [10], where  $p \approx 0.05$  and  $S(0) \approx 0.05$ .

For size disperse systems, we consider not only on  $S(q)$  but also the compressibility,  $\chi_T(q)$ . In matrix notation this reads,

$$[\rho k_B T \chi_T(q)]^{-1} = \mathbf{x}^T \mathbf{S}^{-1}(q) \mathbf{x}, \quad (3)$$

where  $\mathbf{x}^T = (x_1, \dots, x_n)$ . To find  $\chi_T$  one should measure all partial structure factors in  $\mathbf{S}(q)$ , and invert this matrix to get  $\mathbf{S}^{-1}(q)$ . For a one-component (monodisperse) system, one finds  $\rho k_B T \chi_T(q) = S(q)$ , and both quantities are thus fully equivalent. For a binary mixture,  $n = 2$ , one gets [14]

$$\rho k_B T \chi_T(q) = \frac{S_{11}(q)S_{22}(q) - S_{12}^2(q)}{x_1^2 S_{22}(q) + x_2^2 S_{11}(q) - 2x_1 x_2 S_{12}(q)}. \quad (4)$$

In Fig. 1a we show the compressibility measured experimentally. A clear linear behaviour of the compressibility is obtained for low wavevectors. To our knowledge, this anomalous behaviour has not been observed experimentally before. In Fig. 1b we show the compressibility

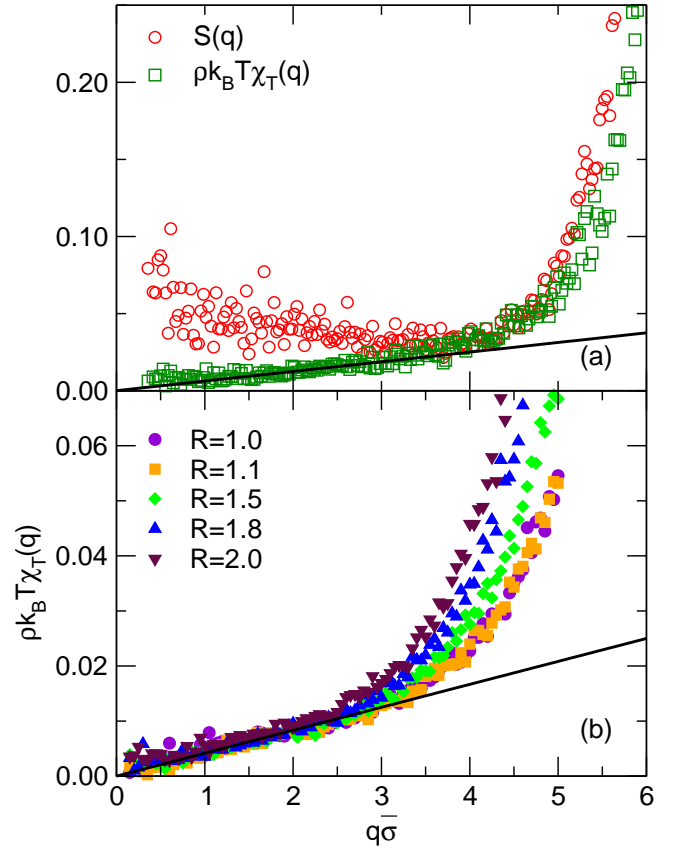


FIG. 1: (a) Structure factor and compressibility for the two-dimensional packings of disks obtained experimentally. While  $S(q)$  resembles that of a binary fluid mixture, the compressibility displays anomalous low- $q$  linear decay. (b) Compressibility for numerically generated jammed packings of 50:50 binary mixtures with various size ratios  $R$ . All systems share the same low- $q$  linear behaviour of the compressibility.

obtained numerically, using Eq. (4), for binary mixtures with different size ratio. For all systems considered, a linear behaviour of the compressibility is obtained for low wavevectors. This set of results suggests that a relevant generalization of Eq. (1) for binary mixtures is obtained by studying  $\chi_T(q)$ , rather than  $S(q)$ .

While straightforward for discrete mixtures with a small number  $n$  of components, where  $\mathbf{S}$  is an  $n \times n$  matrix, the matrix inversion in Eq. (3) is conceptually and computationally difficult for continuous size distributions where the size of the  $\mathbf{S}$ -matrix formally becomes infinite. The system studied experimentally in [10] is of this type. To analyze such packings, we derive a systematic approximation for  $\chi_T(q)$ . The idea is that if the size distribution is sufficiently narrow,  $\chi_T(q)$  can be obtained perturbatively. To this end, we define  $\epsilon_i = (\sigma_i - \bar{\sigma})/\bar{\sigma}$  and derive an expansion of  $\chi_T(q)$  in powers of the  $\epsilon_i$  up to some fixed but otherwise arbitrary order  $a$ . To find a suitable starting point for this expansion, recall the relation  $S_{ij}(q) = x_i \delta_{ij} + \rho x_i x_j h_{ij}(q)$  between the partial structure

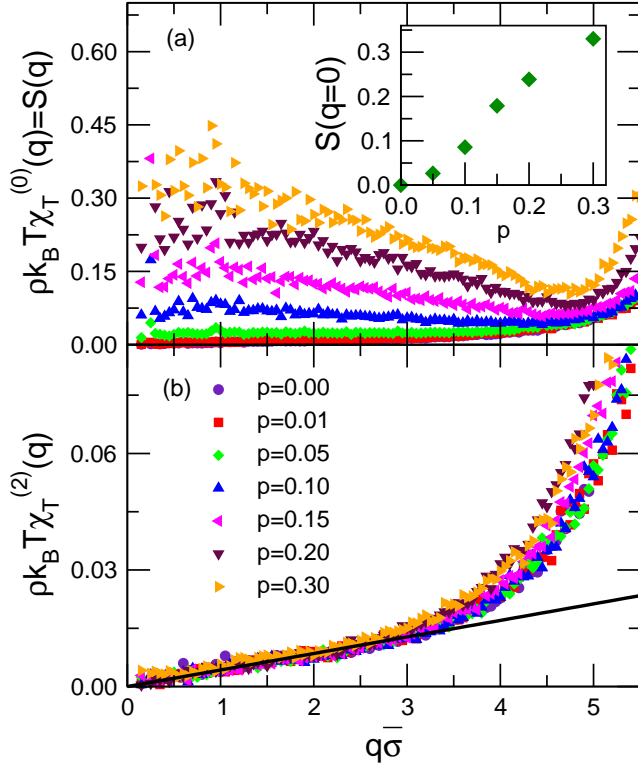


FIG. 2: (a) Zeroth and (b) second order estimate of the compressibility, Eq. (5), for packings with continuous size distribution of polydispersity  $p$ . While the low- $q$  behaviour of the structure factor for  $a = 0$  is featureless (see inset), the compressibility displays an anomalous linear behaviour at low- $q$ , as seen for  $a = 2$ . The second order estimate of the compressibility can be used up to  $p \approx 30\%$ , while the first order one (data not shown) becomes unreliable beyond  $p \approx 10\%$ .

factors and  $h_{ij}(q)$ , the Fourier transforms of the pair correlation functions  $g_{ij}(r) - 1$  [6]. The concentration factors  $x_i$  vary rapidly with  $\epsilon_i$  for narrow distributions, so we

cannot expand  $S_{ij}(q)$ . But the pair correlation functions  $g_{ij}(r)$  and hence the  $h_{ij}(q)$  depend smoothly on particle size, so we expand the latter up to  $\epsilon_i^a \epsilon_j^a$ , taking e.g. for  $a = 1$ ,  $h_{ij}(q) = h_0(q) + h_1(q)(\epsilon_i + \epsilon_j) + h_2(q)\epsilon_i \epsilon_j$  [7]. This expansion is inserted into Eq. (3) for  $S_{ij}(q)$ , giving after some algebra the compact form

$$[\rho k_B T \chi_T^{(a)}(q)]^{-1} = \mathbf{m}_a^T \mathbf{S}_a^{-1}(q) \mathbf{m}_a, \quad (5)$$

where the matrix  $\mathbf{S}_a(q)$  has elements  $S^{\mu\nu}(q) = \frac{1}{N} \langle \epsilon^\mu(\mathbf{q}) \epsilon^\nu(-\mathbf{q}) \rangle$ , with  $\mu, \nu \in \{0, \dots, a\}$ , and captures fluctuations of the moment density fields  $\epsilon^\mu(\mathbf{q}) = \sum_{i=1}^n \epsilon_i^\mu \rho_i(\mathbf{q})$ . Hence,  $\epsilon^0(\mathbf{q}) = \rho(\mathbf{q})$  is the number density field, and  $S^{00}(q) = S(q)$ , the total structure factor. The vector  $\mathbf{m}_a^T = (\delta_0, \dots, \delta_a)$  in Eq. (5) has components given by the moments of  $\epsilon_i$  averaged over the particle size distribution:  $\delta_\mu = \sum_{i=1}^n x_i \epsilon_i^\mu$ , so that  $\delta_0 = 1$ ,  $\delta_1 = 0$ , and  $\delta_2$  is directly related to the polydispersity,  $\delta_2 = p^2$ . The result (5) relates the compressibility to the  $S^{\mu\nu}(q)$  up to order  $a$ . For  $a$  not too large it is simple to compute as it only requires the measurement of  $(a+1)(a+2)/2$  reduced structure factors. It can be applied to arbitrary particle size distributions and is exact for discrete  $n$ -component mixtures if we choose  $a = n - 1$ , as can be shown by direct calculation from Eq. (3).

We have tested our general formula (5) using computer simulations. When  $a = 0$ , one has  $\rho k_B T \chi^{(0)} = S(q)$ , which is only exact for  $n = 1$  (monodisperse systems), as discussed above. At first order,  $a = 1$ , we need to invert a  $(2 \times 2)$  matrix to get  $\rho k_B T \chi_T^{(1)}(q)$ . When applied to the case of a continuous size distribution this formula produces the expected linear behaviour at low  $q$  for  $p \leq 0.10$ , but deviations appear at larger  $p$ . To check whether these deviations are physical, or a result of our approximation, we go to second order,  $a = 2$ , where the required inversion of a  $(3 \times 3)$  matrix gives:

$$\rho k_B T \chi_T^{(2)}(q) = \frac{S^{00} S^{11} S^{22} + 2 S^{02} S^{01} S^{12} - S^{00} [S^{12}]^2 - [S^{01}]^2 S^{22} - [S^{02}]^2 S^{11}}{S^{11} S^{22} - [S^{12}]^2 + 2 \delta_2 (S^{01} S^{12} - S^{02} S^{11}) + \delta_2^2 (S^{00} S^{11} - [S^{01}]^2)}. \quad (6)$$

We now find, see Fig. 2b, that a linear  $q$ -dependence is obtained for polydispersities as large as 30%. This suggests that the same behavior should in fact be obtained for arbitrary size distributions, although measuring the compressibility is more difficult when  $p$  is very large because we need to go to even higher orders  $a$ .

We now discuss the physical significance of our results. It is perhaps not surprising, with hindsight, that jammed sphere packings have vanishing compressibility since this is precisely how the jamming transition was described in

the opening lines of the paper. However, the quantity we call “compressibility” in this work is in fact a particular combination of density fluctuations, Eq. (3), that only reduces to the compressibility at thermal equilibrium when the fluctuation-dissipation theorem holds [6]. Remarkably, our results suggest that a similar connection between response and fluctuations may exist far from equilibrium near jamming, even in real granular packings. An obvious connection between response and fluctuations holds at  $q = 0$ , since  $\chi_T(0) = \phi^{-1} \partial \phi / \partial P$  indeed vanishes

near jamming where  $P \sim (\phi_c - \phi)^{-1}$ , so that both sides of Eq. (3) vanish. It would be interesting to extend these considerations to finite  $q$  near the jamming transition.

Our results also illustrate that, for size disperse systems, the limit  $S(q \rightarrow 0)$  is in general not directly related to the isothermal compressibility,  $\chi_T(0)$  [15]. While the latter vanishes in jammed packings, the former is free to take any positive value. This explains why previous work on size disperse packings failed to observe any anomalous behaviour [9, 10]. Both quantities are related to the amplitude of fluctuations of the number density, but  $S(q \rightarrow 0) \sim \langle \Delta N^2 \rangle$  captures the total fluctuation of  $N$  while  $\chi_T(q \rightarrow 0)$  quantifies the fluctuations of  $N$  at fixed composition. This can be seen by defining the composition fluctuation fields  $c_i(\mathbf{q}) = \rho_i(\mathbf{q}) - x_i \rho(\mathbf{q})$  for  $i = 1, \dots, n - 1$ . The compressibility from Eq. (3) can then be rewritten as

$$\rho k_B T \chi_T(q) = S(q) - \mathbf{s}_{0c}^T(q) \mathbf{S}_{cc}^{-1}(q) \mathbf{s}_{0c}(q) \quad (7)$$

where the  $(n - 1)$ -dimensional vector  $\mathbf{s}_{0c}$  gathers the correlations between number and composition fluctuations, and the matrix  $\mathbf{S}_{cc}$  the correlations among the latter [14, 16]. For the compressibility to vanish at jamming the two terms must cancel, which implies that local fluctuations in  $N$  become fully correlated with composition fluctuations. On the other hand,  $S(q \rightarrow 0)$  remains positive because a local fluctuation of  $N$  can be induced by a local fluctuation of the mixture composition: fluctuations of  $N$  do occur in jammed *size disperse* packings.

The behaviour of  $S(q)$  in Fig. 2 is clearly inconsistent with Eq. (1), as noticed previously [9, 10]. Does this imply that jammed size disperse packings are not hyperuniform, as suggested in [9]? For point particles, hyperuniformity refers to vanishing bulk fluctuations of the number density, as described in the introduction. However, for an assembly of spherical particles hyperuniformity requires vanishing bulk fluctuations of the local volume fraction [17]. While  $N$  and  $\phi$  are directly proportional for monodisperse spheres like those studied in Ref. [4], they are not when the packing is size disperse, and thus no conclusion can be drawn from  $S(q)$  alone.

A connection between the anomalous compressibility studied in this work and suppressed fluctuations of the volume fraction in hyperuniform packings can be established. From Eq. (3), we realize that whenever the  $\mathbf{S}(q)$  matrix possesses at least one eigenvalue that goes to zero at low  $q$ , the compressibility vanishes. We have diagonalized  $\mathbf{S}(q)$  or  $\mathbf{S}_a(q)$  as obtained in our numerical and experimental packings, and indeed found that in each case anomalous behaviour of the compressibility originates from a single vanishing eigenvalue. This implies that there exists a particular linear combination of the partial density fields which has no bulk fluctuations. A detailed analysis of the corresponding eigenvectors shows that they are fully compatible with the local definition of the volume fraction,  $\phi(\mathbf{q}) = (\pi/6) \sum_{i=1}^n \sigma_i^d \rho_i(\mathbf{q})$ . This

identification holds exactly for binary mixtures in our  $3d$  simulations and in the  $2d$  experimental packings. It also holds true for the packings with continuous size distributions, to the same order in  $\epsilon_i$  that we analyse for the compressibility ( $a = 2$ , which is accurate up to polydispersity  $p = 30\%$ ). Indeed, we have checked that direct measurements of  $I(q) = \langle \phi(\mathbf{q}) \phi(-\mathbf{q}) \rangle$  coincide with the compressibility shown throughout this article in the low- $q$  regime where linear behaviour is observed. Therefore, the anomalous behaviour of the compressibility reveals the absence of bulk fluctuations of the volume fraction. This lack of fluctuations was detected also in the recent, independent, study of Ref. [11], using a rather different methodology. We conclude that all our size disperse jammed packings are hyperuniform.

We have demonstrated anomalous behaviour of the compressibility in jammed size disperse packings of spheres both in simulations and in a granular experiment, using for the case of continuous size distributions an efficient perturbative approach. We have related this to suppressed bulk fluctuations of the volume fraction, or hyperuniformity (see also [11], thus revealing a structural signature of jamming not seen in the conventional structure factor. Our work also raises intriguing questions about the applicability of fluctuation-dissipation relations to jammed systems.

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